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Predictive Modeling and Experimental Characterization of Non-Thermal Plasma Treatment of Gaseous Emissions

B. M. Penetrante, M. C. Hsiao, B. T. Merritt,
G. E. Vogtlin, and P. H. Wallman
Lawrence Livermore National Laboratory

A. Kuthi, C. P. Burkhart, and J. R. Bayless
First Point Scientific, Inc.

The electron mean energy in a plasma reactor is very important because it determines the types of radicals produced in the plasma and the input electrical energy required to produce those radicals. Electron mean energies around 5 eV are optimum for the electron-impact dissociation of O_2, which is important for the production of O radicals and ozone. These oxidizing radicals play a key role in the initial decomposition of some types of volatile organic compounds (VOCs). To implement the chemical reduction of NO to benign molecules such as N_2 and O_2, the important radical is the N radical, which is produced through the electron-impact dissociation of N_2. High electron mean energies (>10 eV) are required to efficiently implement the dissociation of N_2. For processes requiring OH radicals, high electron mean energies are also more efficient because the positive ions produced by ionization processes lead to OH production, more than can be achieved by direct dissociation of H_2O by either electrons or $O(^1D)$. For some VOCs like CCl_4, for which the initial decomposition goes through electron attachment to thermal electrons, high incident electron energies provide a more efficient means (higher G values) for producing copious amounts of electrons in the plasma.

Most electrical discharge reactors produce a non-thermal plasma through the formation of microdischarges known as streamers. During the streamer formation phase, the electron mean energy at the streamer head reaches values of more than 10 eV - suitable for large dissociation and ionization of the gas. However, since this is a highly transient phase, and since the ionization wave covers only small parts of the gap at the same time, this phase seems to be less important in producing most of the active radicals. The microdischarge column during the main current flow is the region where the main portion of radicals is generated. This column covers a large part of the gap and lasts throughout the lifetime of a microdischarge. Thus, most of the radicals are produced in the streamer channel after the streamer head has crossed the electrode gap. The streamer plasma is space-charge shielded; the streamer channel field dictates the electron mean energy that determines the radical production efficiency of the plasma reactor. For atmospheric-pressure air discharges, the channel field corresponds to electron mean energies of 3 to 4 eV. Because of space-charge shielding, the electron mean energy in the plasma channels weakly depends on the electrode structure, applied peak voltage, and whether the microdischarges are extinguished because of the short voltage pulse (as in pulsed corona) or because of the dielectric charging (as in dielectric-barrier discharge).

Figure 1 shows a comparison of electron beam, pulsed corona and dielectric-barrier discharge processing of 100 ppm NO in N_2. Note that electron beam processing is six times more energy efficient than either pulsed corona or dielectric-barrier discharge processing. In real exhausts containing N_2, O_2, CO_2, H_2O and other dilute components, the only process that can lead to NO chemical reduction is the electron-impact dissociation of N_2. By performing experiments using dilute mixtures of NO in N_2, as in Figure 1, we have been able to measure precisely the amount of N_2 dissociation that can be achieved in different types of plasma reactors. Our low-voltage electron beam experiments show excellent agreement with electron beam experiments at the Japan Atomic Energy Research Institute (JAERI), which used a 1.5 Megavolt electron accelerator. This is as ex-

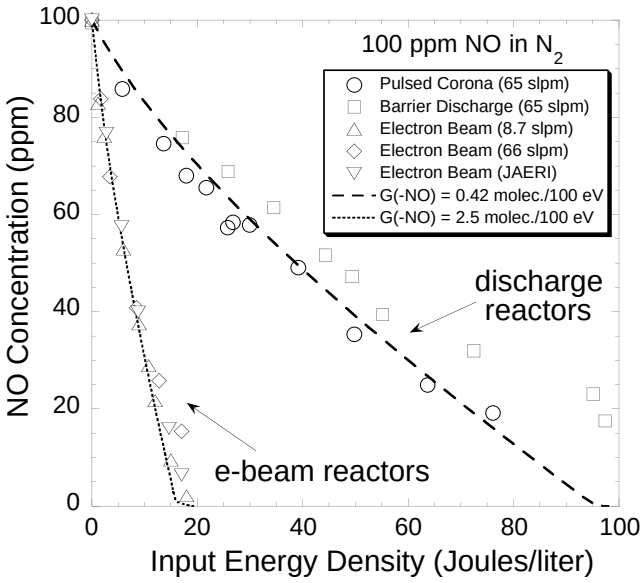

Figure 1

pected, since the ratios of the electron-impact cross sections are constant for incident electron energies above 100 eV. Our experiments demonstrated that compact electron beam processing is possible because we do not have to rely on big and expensive electron accelerators to achieve high electron mean energies in the created plasma.

We are also conducting studies on the plasma assisted decomposition of VOCs in atmospheric-pressure air streams. Figure 2 shows a comparison between pulsed corona and dielectric-barrier discharge processing of trichloroethylene (TCE). The experiments were performed at gas temperatures up to 300°C. One of the objectives in these experiments is to study the effect of gas temperature on the removal chemistry and product formation. The data on the gas temperature dependence provide a good basis for elucidating the chemical kinetics of TCE decomposition in the plasma. Under identical gas conditions (i.e. gas composition and gas temperature), the type of electrical discharge reactor does not affect the electrical energy requirements for decomposing the same amount of TCE; the reactor type also does not affect the product formation. For input energy densities up to 300 Joules per liter, we observe that CO and CO_2 are only minor products in the decomposition of TCE. The main organic products are phosgene and dichloroacetyl chloride (DCAC), as observed from the FTIR spectra of the effluent. Processing at higher gas temperatures (around 300°C) increases the electrical energy required to remove the same amount of TCE; however, the CO and CO_2 yields increase substantially and higher amounts of HCl are formed. These trends suggest increased competition from decomposition of DCAC and/or phosgene at high temperatures. In all cases, pulsed corona or dielectric-barrier discharge processing produces CO preferentially over CO_2.

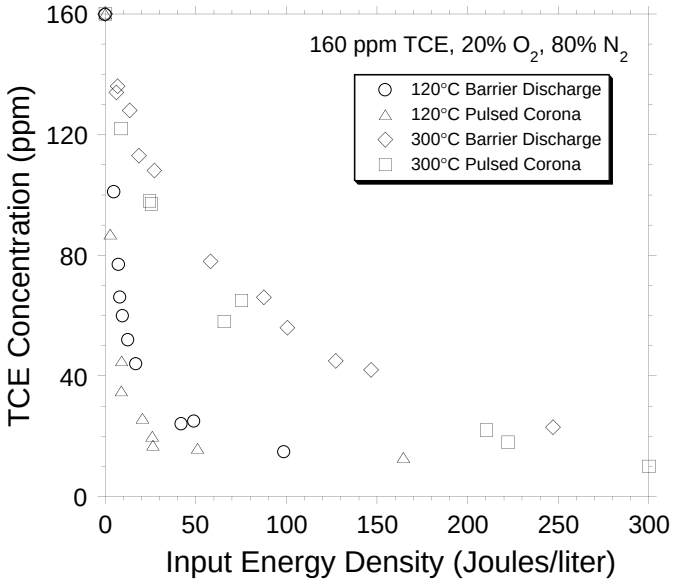

Figure 2

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